

# Limited influence of diluted ferromagnetic dimers on Curie temperature in complex magnetic systems.

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In this work we investigate Ising and classical Heisenberg models for two and three dimensional lattices in presence of diluted ferromagnetic dimers. For such models the Curie temperature as a function of ratio of intra-dimer exchange coupling constant  $I_A$  and other inter-site coupling constants  $I_B$  is calculated. In case dimer is treated exactly and environment within the mean-field approach it was found that even for  $I_A/I_B \rightarrow \infty$   $T_C$  remains finite. Similar analysis is proposed for rhombohedral phase of intermetallic compound  $\text{Gd}_2\text{Fe}_{17}$  where so-called Fe1-Fe1 “dumbbell” forms the diluted ferromagnetic dimer. It was shown that for such complex magnetic systems  $T_C$  is determined by all variety of exchange interactions and for the interval  $0 \leq I_A/I_B \leq \infty$   $T_C$  changes are not more than  $\pm 10\%$ .

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## I. INTRODUCTION

There is a number of magnetic compounds with complex crystal structure and many magnetic ions per unit cell. Interatomic distances in such systems between nearest exchange-coupled ionic pairs could be quite different by its value. Thus the value of exchange coupling parameter for a pair with shortest interatomic distance could be 2-3 times stronger than for a pair of atoms with larger interatomic distance.

A typical example of such systems is  $\text{R}_2\text{Fe}_{17}$  class of intermetallic compounds. Here shortest Fe1-Fe1 distance corresponds to Fe ions in a so called “dumbbell” positions (Wyckoff positions 4f for hexagonal  $\text{Th}_2\text{Ni}_{17}$ -type and 6c for rhombohedral  $\text{Th}_2\text{Zn}_{17}$ -type crystal structures, see for details of magnetic structure Ref. 1). Based on band structure calculations within the LSDA+U method the exchange interaction parameter value for dumbbell pair in the hexagonal phase  $\text{Gd}_2\text{Fe}_{17}$  gives ferromagnetic exchange with  $I_{11}(1)=238.8$  K for a distance  $r_{11}(1)=2.400\text{\AA}$ .<sup>2</sup> While for Fe3-Fe3 pairs in the 12j Wyckoff positions for a distance of  $r_{33}(1)=2.400$  Å corresponding exchange parameter value is found to be  $I_{33}(1)=80.4$  K.<sup>2</sup> Analogous calculations for rhombohedral phase of  $\text{Gd}_2\text{Fe}_{17}$  for Fe1-Fe1 dumbbell pair gives ferromagnetic coupling with  $I_{11}(1)=287.5$  K ( $r_{11}(1)=2.385$  Å).<sup>2</sup> For the next pair interatomic distance Fe2-Fe3 (9d and 18f Wyckoff positions correspondingly) we obtained  $I_{23}(1)=87.1$  K ( $r_{23}(1)=2.423$  Å)<sup>2</sup>. Thus one can see that coupling for dumbbell positions is 3 times stronger (in both phases) than an exchange coupling for the next smallest pair interatomic Fe-Fe distance.

Let us note that these strong exchange bonds  $I_{11}(1)$  of Fe1-Fe1 dumbbell positions are rather well spatially separated one from each other (rather diluted) and do not form any infinite magnetic cluster. Moreover these strongly coupled dumbbell pairs one can consider as some ferromagnetic dimers embedded into a infinite magnetic

cluster formed by other weaker magnetic bonds.

To this end there rises a question: how strong influences presence of such diluted ferromagnetic dimers on a Curie temperature? Well could be that such dimers give dominating contribution to the Curie temperature of a complex magnetic system described above.

Here we solve a problem of influence of diluted ferromagnetic dimers on a Curie point. First for model lattices with Ising or classical Haisenberg spins with different numbers of lattice sites we vary exchange interaction strengths of the dimer from zero to infinity while all other exchange interactions remains finite (see Sec. II). Then we investigate rhombohedral phase of  $\text{Gd}_2\text{Fe}_{17}$  using the same approach (Sec. III). Finally we conclude our work with a summary (Sec. IV).

## II. THE CURIE TEMPERATURE FOR MODEL LATTICES WITH DILUTED FERROMAGNETIC DIMERS

### A. Ising spins case

Let us consider square lattice with lattice parameter  $a$ . On each site we have Ising spin  $\tau_i = \pm 1$ . We also specify that spins  $\tau_1$  and  $\tau_2$  on a plaquette (selected on Fig. 1 by dashed circle) form a dimer and are coupled by exchange integral  $I_A > 0$  while each other pair of spins  $\tau_1 - \tau_3$ ,  $\tau_3 - \tau_4$  and  $\tau_4 - \tau_2$  are coupled with  $I_B > 0$  and  $I_B \neq I_A$ . Thus magnetic elementary cell contains four magnetic atoms and the lattice has period  $2a$ .

Using Weiss mean-field approximation it is easy to obtain four self-consistent equations for observable spin value  $\sigma_i = \langle \tau_i \rangle$  ( $i=1,2,3,4$ ) for each magnetic sublattice.

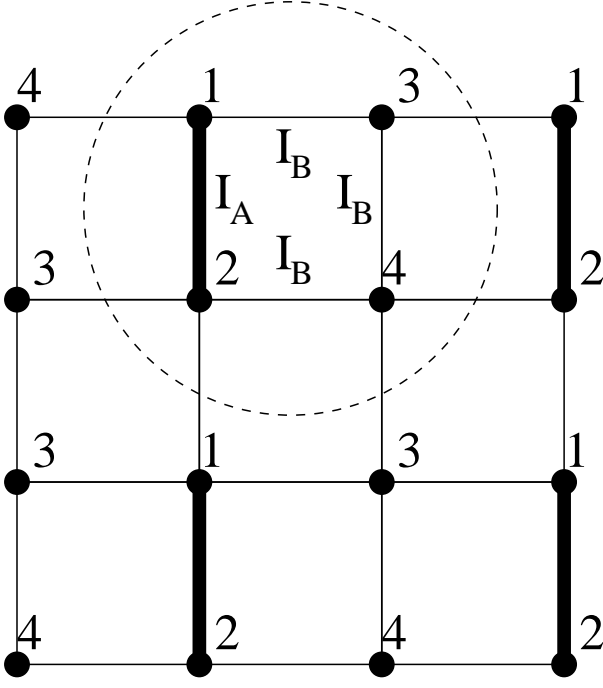


FIG. 1: Square lattice with spins in the lattice sites with the period  $2a$  (magnetic unit cell surrounded by dashed circle). Strong exchange bond  $I_A$  — fat black line, thick lines — exchange bonds  $I_B$ . Numbers 1,2,3,4 enumerate spins (magnetic sublattices) in the magnetic unit cell.

$$\begin{aligned}
 \sigma_1 &= th\{\beta[I_A\sigma_2 + I_B(\sigma_2 + 2\sigma_3)]\} \\
 \sigma_2 &= th\{\beta[I_A\sigma_1 + I_B(\sigma_1 + 2\sigma_4)]\} \\
 \sigma_3 &= th\{\beta I_B(2\sigma_1 + 2\sigma_4)\} \\
 \sigma_4 &= th\{\beta I_B(2\sigma_2 + 2\sigma_3)\}
 \end{aligned} \quad (1)$$

with  $\beta = 1/k_B T$ .

If  $T \rightarrow T_C$  and  $\sigma_1, \sigma_2, \sigma_3, \sigma_4 \rightarrow 0$  Eq. 1 reduces to linear equations determining  $T_C$

$$\begin{aligned}
 \sigma_1 &= \beta_C[(I_A + I_B)\sigma_2 + 2I_B\sigma_3] \\
 \sigma_2 &= \beta_C[(I_A + I_B)\sigma_1 + 2I_B\sigma_4] \\
 \sigma_3 &= \beta_C I_B(2\sigma_1 + 2\sigma_4) \\
 \sigma_4 &= \beta_C I_B(2\sigma_2 + 2\sigma_3).
 \end{aligned} \quad (2)$$

For ferromagnetic ordering in elementary magnetic cell shown on Fig. 1 follows relations  $\sigma_1 = \sigma_2$  and  $\sigma_3 = \sigma_4$ . Then

$$k_B T_C = \frac{1}{2} \left( I_A + 3I_B + \sqrt{I_A^2 - 2I_A I_B + 17I_B^2} \right) \quad (3)$$

If  $I_A = 0$  it comes out from Eq. (3)  $k_B T_C(I_A = 0) = \frac{1}{2}(3 + \sqrt{17})I_B = 3.562I_B$  and for  $I_A = I_B$  one can find

$k_B T_C(I_A = I_B) = 4I_B$ . However in limiting case  $I_A \rightarrow \infty$  Eq. (3) leads to  $k_B T_C \rightarrow \infty$ .

It is obvious that the above mentioned result is not correct from physical point of view. In case  $I_A \gg I_B$  strong ferromagnetic exchange coupling  $I_A$  within a dimer preserves parallel alignment of spins  $\tau_1$  and  $\tau_2$ . While decoupling of weak exchange bonds  $I_B$  (which form infinite ferromagnetic cluster) is responsible for destruction of ferromagnetic order. Thus even for  $I_A \rightarrow \infty$  Curie temperature  $T_C$  should be finite and proportional to  $I_B$ .

The origin of such paradox is applied mean-field approximation where strong fluctuations of exchange energy because of spin flips are absent. The fluctuations become anomalously strong if we deal with groups of spins strongly coupled with each other. To this end one should use approach proposed by Scalapino, Imry and Pincus<sup>3</sup> for quasi one dimensional magnetic systems with two significantly different exchange constants  $I_A$  and  $I_B$ . The main idea is to treat strongly coupled spins exactly while weak exchange interactions are treated within the mean-field approximation.

For this purpose let us write down a Hamiltonian of such two spins cluster (dimer) in the elementary magnetic cell shown on Fig. 1

$$\begin{aligned}
 \mathbf{H}_{cl} &= -I_A \tau_1 \tau_2 - I_B(\sigma_2 + 2\sigma_3)\tau_1 - \\
 &\quad - I_B(\sigma_1 + 2\sigma_4)\tau_2.
 \end{aligned} \quad (4)$$

Here interaction between spins  $\tau_1$  and  $\tau_2$  in the dimer which are coupled with a large exchange constant  $I_A$  is kept in the exact form and interaction of the same spins  $\tau_1$  and  $\tau_2$  with others via small  $I_B$  is treated within the mean-field approximation.

Partition function for the dimer in the environment is

$$\begin{aligned}
 Z_{cl} &= 2 \left\{ e^{\beta I_A} ch[\beta I_B(\sigma_1 + \sigma_2 + 2\sigma_3 + 2\sigma_4)] + \right. \\
 &\quad \left. + e^{-\beta I_A} ch[\beta I_B(\sigma_1 - \sigma_2 - 2\sigma_3 + 2\sigma_4)] \right\}.
 \end{aligned} \quad (5)$$

Then for average values of spins  $\sigma_1 = \langle \tau_1 \rangle$  and  $\sigma_2 = \langle \tau_2 \rangle$  one can derive self-consistent set of equations

$$\begin{aligned}
 \sigma_1 &= \frac{2}{Z_{cl}} \left\{ e^{\beta I_A} sh[\beta I_B(\sigma_1 + \sigma_2 + 2\sigma_3 + 2\sigma_4)] - \right. \\
 &\quad \left. - e^{-\beta I_A} sh[\beta I_B(\sigma_1 - \sigma_2 - 2\sigma_3 + 2\sigma_4)] \right\},
 \end{aligned} \quad (6)$$

$$\begin{aligned}
 \sigma_2 &= \frac{2}{Z_{cl}} \left\{ e^{\beta I_A} sh[\beta I_B(\sigma_1 + \sigma_2 + 2\sigma_3 + 2\sigma_4)] + \right. \\
 &\quad \left. + e^{-\beta I_A} sh[\beta I_B(\sigma_1 - \sigma_2 - 2\sigma_3 + 2\sigma_4)] \right\}.
 \end{aligned} \quad (7)$$

For average values of spins  $\sigma_3 = \langle \tau_3 \rangle$  and  $\sigma_4 = \langle \tau_4 \rangle$  weakly coupled to environment via  $I_B$  still valid last two equations of Eqs. (1).

It is remarkable that for the limit  $I_A \rightarrow \infty$  immediately follows

$$\sigma_1 = \sigma_2 = th[\beta I_B(\sigma_1 + \sigma_2 + 2\sigma_3 + 2\sigma_4)]. \quad (8)$$

It means that for infinitely strong exchange coupling between spins  $\sigma_1 = \langle \tau_1 \rangle$  and  $\sigma_2 = \langle \tau_2 \rangle$  (in case the spins are strictly parallel to each other) environment mean-field  $I_B(\sigma_1 + 2\sigma_4)$  acting on spin  $\tau_2$  which belongs to the dimer is transferred onto spin  $\tau_1$  and thus is added to Weiss field  $I_B(\sigma_2 + 2\sigma_3)$ . Similar “transfer” of Weiss field from spin  $\tau_1$  happens to spin  $\tau_2$ .

If we turn back to finite values of  $I_A$ , considering  $\sigma_1 = \sigma_2$  and  $\sigma_3 = \sigma_4$  because of symmetry of magnetic surrounding, one can obtain two self-consistent equations

$$\begin{aligned}\sigma_1 &= \frac{sh[\beta I_B(2\sigma_1 + 4\sigma_3)]}{ch[\beta I_B(2\sigma_1 + 4\sigma_3)] + e^{-2\beta I_A}}, \\ \sigma_3 &= th[2\beta I_B(\sigma_1 + \sigma_3)].\end{aligned}\quad (9)$$

In case of  $T \rightarrow T_C$  one can linearize Eqs. (9) for  $\sigma_1, \sigma_3 \rightarrow 0$  and get uniform linear set of equations

$$\begin{aligned}\sigma_1 &= \beta_C I_B [1 + th(\beta_C I_A)](\sigma_1 + 2\sigma_3), \\ \sigma_3 &= 2\beta_C I_B(\sigma_1 + \sigma_3).\end{aligned}\quad (10)$$

Since Eqs. (10) are valid only for  $I_A \gg I_B$ , then for  $I_A = 10I_B$  one can find  $k_B T_C(I_A = 10I_B) = 4.792I_B$  and for  $I_A \rightarrow \infty$  correspondingly  $k_B T_C(I_A \rightarrow \infty) = 2(1 + \sqrt{2})I_B = 4.828I_B$ .

To summarize this subsection we have investigated a square lattice model with four spins in the elementary magnetic cell and one strong exchange bond  $I_A$  out of four. Varying the selected exchange coupling constant in the interval  $0 < I_A < \infty$  we obtained variation of the Curie point from  $k_B T_C(I_A = 0) = 3.562I_B$  upto  $k_B T_C(I_A \rightarrow \infty) = 4.828I_B$ .

### B. Elementary magnetic cell finite size effect on $T_C$

Now one traces how an increase of number of magnetic sites in the elementary magnetic cell changes Curie temperature in presense of the dimer. In case of square lattice with the periodicity of magnetic elementary cell equal to  $3a$  containing nine magnetic sites (see Fig. 2) magnetic structure is described by nine magnetic sublattices. However using symmetry of magnetic environment one can find following relations between different sublattice spin values  $\sigma_1 = \sigma_2$ ,  $\sigma_4 = \sigma_5 = \sigma_7 = \sigma_8$  and  $\sigma_6 = \sigma_9$ . Thus if  $0 \leq I_A \lesssim I_B$  employing mean-field approximation for  $T \rightarrow T_C$  one can obtain four linear equations

$$\begin{aligned}\sigma_1 &= \beta_C [I_A \sigma_1 + I_B(\sigma_3 + 2\sigma_4)], \\ \sigma_3 &= \beta_C [I_B(2\sigma_1 + 2\sigma_6)], \\ \sigma_4 &= \beta_C [I_B(\sigma_1 + 2\sigma_4 + \sigma_6)], \\ \sigma_6 &= \beta_C [I_B(\sigma_3 + 2\sigma_4 + \sigma_6)].\end{aligned}\quad (11)$$

By solving fourth order determinant one can get  $k_B T_C(I_A = 0) = 3.820I_B$ .

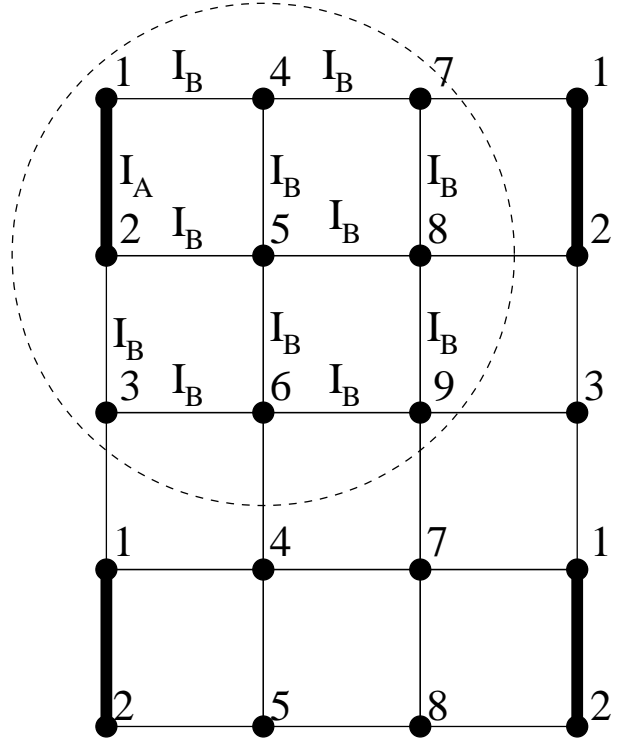


FIG. 2: The same as Fig. 1, but for the period  $3a$  (9 magnetic sublattices).

In case of strong exchange bond  $I_A \gg I_B$  it is necessary to write corresponding Hamiltonian similar to Eq. (4) using relations  $\sigma_4 = \sigma_5 = \sigma_7 = \sigma_8$

$$\begin{aligned}\mathbf{H}_{cl}(1, 2) &= -I_A \tau_1 \tau_2 - I_B(\sigma_3 + \sigma_4 + \sigma_7) \tau_1 - \\ &\quad - I_B(\sigma_3 + \sigma_5 + \sigma_8) \tau_2 = \\ &= -I_A \tau_1 \tau_2 - I_B(\sigma_3 + 2\sigma_4)(\tau_1 + \tau_2).\end{aligned}\quad (12)$$

Then average value  $\sigma_1 = \langle \tau_1 \rangle$  is

$$\sigma_1 = \frac{sh[\beta I_B(2\sigma_3 + 4\sigma_4)]}{ch[\beta I_B(2\sigma_3 + 4\sigma_4)] + e^{-2\beta I_A}}.\quad (13)$$

In the vicinity of  $T_C$  Eq. (13) is linearized for small values  $\sigma_3, \sigma_4 \rightarrow 0$  and gives

$$\sigma_1 = \beta_C I_B [1 + th(\beta_C I_A)](\sigma_3 + 2\sigma_4),\quad (14)$$

and should be used as a first equation in the Eqs. (11). Then one can calculate  $k_B T_C(I_A = 10I_B) = 4.331I_B$  and  $k_B T_C(I_A \rightarrow \infty) = 4.340I_B$ .

Similar calculations were done by us also for the case of two dimensional lattice with period  $4a$  as well as for three dimensional models with elementary magnetic cells ( $2a \times 2a \times 2a$ ) shown on Fig. 3 (8 magnetic sublattices), ( $3a \times 3a \times 2a$ ) (18 magnetic sublattices) and ( $3a \times 3a \times 3a$ ) (27 magnetic sublattices). Corresponding results are collected in Table I. It is clearly seen that under increase of number of magnetic sites in the elementary magnetic

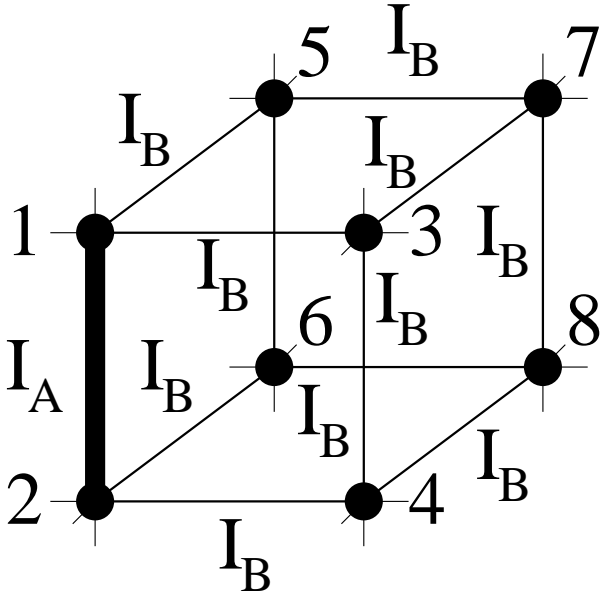


FIG. 3: The same as Fig. 1, but for the three dimensional cubic lattice (8 magnetic sublattices).

cell and corresponding increase of number of weak magnetic bonds influence of the dimer on  $T_C$  goes down and becomes insignificant even in the limit  $I_A \rightarrow \infty$ .

Roughly speaking infinitely strong exchange bond  $I_A$  (in case spins are strictly parallel in the dimer) produces an effective spin with doubled value of magnetic moment. At the same time it decreases number of magnetic sites by one. After all such dimer contributes rather weak into free energy of the system in comparison with other spins. The same data is displayed on Fig. 4. Here one can see a tendency for the  $T_C$  to lesser and lesser deviate from  $T_C(I_A = I_B)$  for larger number of atoms in the elementary magnetic cell.

### C. Classical Heisenberg spins case

In our days there is a way to compute exchange interaction parameters between different sites of Heisenberg model with classical spins proposed by Lichtenstein *et al.* in Ref. 4. In this approach exchange parameters are calculated as a second derivative of total energy with respect to small angles of magnetic moments deviation from collinear magnetic configuration. Thus one should understand how Curie temperature of ferromagnetic lattice of classical Heisenberg spins depends on presence of diluted magnetic dimers. Here we present analysis similar to Sec. II A.

At the beginning we consider a lattice shown on Fig. 1 with classical spin vectors  $\vec{S}_i = S(\sin\theta_i \cos\phi_i, \sin\theta_i \sin\phi_i, \cos\theta_i)$ . Now we calculate average values of spin vectors for ferromagnetic case assuming that spontaneous magnetic moment is directed

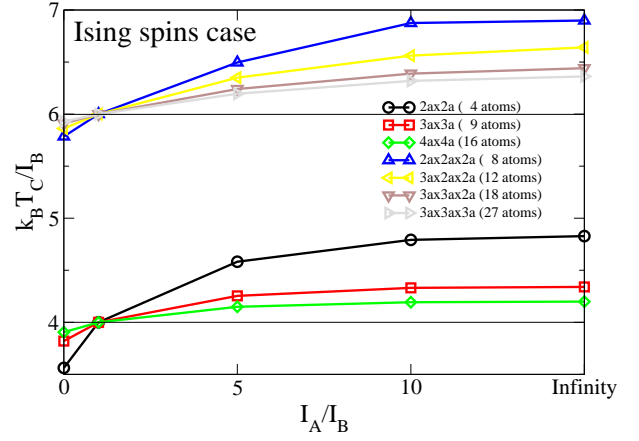


FIG. 4: Dimensionless Curie temperature versus selected exchange bond strength to other exchange bond strengths ratio  $I_A/I_B$  for two and three dimensional cubic lattices of Ising spins with different size of elementary magnetic cells.

along  $z$ -axis. It means that  $\langle S_i^x \rangle = \langle S_i^y \rangle = 0$  and  $m_i \equiv \langle S_i^z \rangle = S \langle \cos\theta_i \rangle \neq 0$ . Within the mean-field approximation for spins average value on sites of each of four magnetic sublattices one can get following equations:

$$\begin{aligned} m_1 &= S\mathcal{L}(\beta S[I_A m_2 + I_B(m_2 + 2m_3)]), \\ m_2 &= S\mathcal{L}(\beta S[I_A m_1 + I_B(m_1 + 2m_4)]), \\ m_3 &= S\mathcal{L}(\beta S I_B[2m_1 + 2m_4]), \\ m_4 &= S\mathcal{L}(\beta S I_B[2m_2 + 2m_3]). \end{aligned} \quad (15)$$

where  $\mathcal{L}(x) = \text{cth}(x) - \frac{1}{x}$  - Langevin function. For  $T \rightarrow T_C$   $\mathcal{L}(x) \approx \frac{x}{3}$ ,  $x \ll 1$  (compare with Eq. 2):

$$\begin{aligned} m_1 &= \beta_C \frac{S^2}{3} [(I_A + I_B)m_2 + 2I_B m_3], \\ m_2 &= \beta_C \frac{S^2}{3} [(I_A + I_B)m_1 + 2I_B m_4], \\ m_3 &= \beta_C \frac{S^2}{3} I_B(2m_1 + 2m_4), \\ m_4 &= \beta_C \frac{S^2}{3} I_B(2m_2 + 2m_3). \end{aligned} \quad (16)$$

Exploiting symmetry relations  $m_1 = m_2$  and  $m_3 = m_4$  one can end up with equation on  $T_C$

$$k_B T_C = \frac{S^2}{3} \frac{1}{2} \left( I_A + 3I_B + \sqrt{I_A^2 - 2I_A I_B + 17I_B^2} \right), \quad (17)$$

which differs from Eq. (3) only with coefficient  $\frac{S^2}{3}$  on the left side. Similar to Ising case simply mean-field treatment gives incorrect results, for the case  $\frac{I_A}{I_B} \rightarrow \infty$  namely,  $T_C \rightarrow \infty$ .

Let us employ once more the approach of Ref. 3, where for the case  $I_A \gg I_B$  for a dimer we write down a Hamiltonian with exchange interaction in the dimer is taken

TABLE I: Curie temperature value for different magnetic elementary cells with Ising spins with selected strong exchange bond  $I_A$  as a function of ratio  $I_A/I_B$ .  $T_C$  is given in units  $I_B$ .

Magnetic elementary cell	Magnetic atoms number	$I_A=0$	$I_A=I_B$	$I_A=5I_B$	$I_A=10I_B$	$I_A \rightarrow \infty$
$2a \times 2a$	4	3.562	4	4.582	4.792	4.828
$3a \times 3a$	9	3.820	4	4.254	4.331	4.340
$4a \times 4a$	16	3.905	4	4.149	4.193	4.199
$2a \times 2a \times 2a$	8	5.785	6	6.496	6.785	6.899
$3a \times 2a \times 2a$	12	5.862	6	6.350	6.562	6.641
$3a \times 3a \times 2a$	18	5.910	6	6.241	6.389	6.441
$3a \times 3a \times 3a$	27	5.929	6	6.196	6.319	6.362

exactly. The exchange coupling constant is  $I_A$  and spin vectors  $\vec{S}_1, \vec{S}_2$ . Coupling of these two spins to other ones via weak exchange integral  $I_B$  we consider within mean-field approximation:

$$\mathbf{H}_{cl}(1, 2) = -I_A \vec{S}_1 \vec{S}_2 - h_1 (S_1^z + S_2^z), \quad (18)$$

here Weiss field is  $h_1 = I_B(m_1 + 2m_3)$ . Corresponding partition function is

$$\begin{aligned} Z_{cl}(1, 2) &= \int d\Omega_1 \int d\Omega_2 \exp\{\beta[I_A \vec{S}_1 \vec{S}_2 + h_1(S_1^z + S_2^z)]\} = \\ &= 4\pi \int d\Omega_1 \exp(\beta S h_1 \cos\theta_1) \times \\ &\times \frac{sh(\beta S \sqrt{I_A^2 S^2 + 2I_A S h_1 \cos\theta_1 + h_1^2})}{\beta S \sqrt{I_A^2 S^2 + 2I_A S h_1 \cos\theta_1 + h_1^2}}, \quad (19) \end{aligned}$$

where  $d\Omega_i = \sin\theta_i d\theta_i d\phi_i$  is solid angle element with  $0 < \theta_i < \pi$ ,  $0 < \phi_i < 2\pi$ .

Since to get  $T_C$  one needs to linearize equations for  $m_1$  and  $m_3$  (that is  $h_1$ ). Therefore we calculate  $Z_{cl}(1, 2)$  up to the order  $h_1^2$ .

$$\begin{aligned} Z_{cl}(1, 2) &= (4\pi)^2 \frac{sh(\beta I_A S^2)}{\beta I_A S^2} \{1 + \\ &+ \frac{1}{3}[1 + \mathcal{L}(\beta I_A S^2)](\beta S h_1)^2 + \dots\}. \quad (20) \end{aligned}$$

Then average value of spin  $m_1$  on site 1 in the elementary magnetic cell is

$$\begin{aligned} m_1 &= \frac{1}{2}(m_1 + m_2) = \frac{1}{2}(\langle S_1^z \rangle + \langle S_2^z \rangle) = \\ &= \frac{1}{2} \frac{1}{Z_{cl}(1, 2)} \frac{\partial}{\partial(\beta h_1)} Z_{cl}(1, 2). \end{aligned}$$

After differentiating of Eq. (20) and following expanding of self-consistent equation for  $m_3$  around small values of  $m_1$  and  $m_3$  we come to two equations

$$\begin{aligned} m_1 &= \beta_C I_B \frac{S^2}{3} [1 + \mathcal{L}(\beta_C I_A S^2)] (m_1 + 2m_3), \\ m_3 &= 2\beta_C I_B \frac{S^2}{3} (m_1 + m_3). \quad (21) \end{aligned}$$

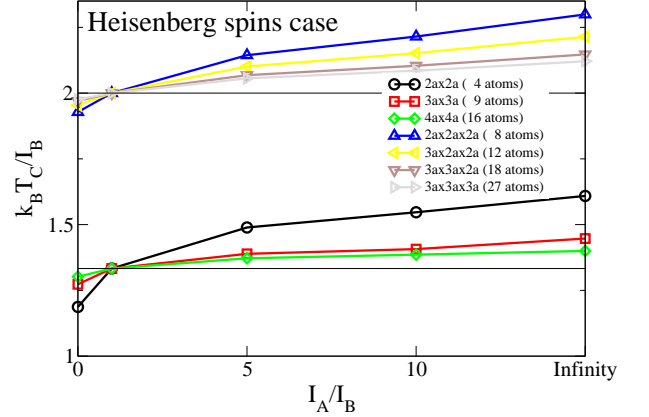


FIG. 5: The same as Fig. 4, but for classical Heisenberg spins.

Last equations for  $T_C$  coincides with Eqs. (10) for Ising spins upto substitutions  $I_B \frac{S^2}{3} \rightarrow I_B$  and  $\mathcal{L}(\beta_C I_A S^2) \rightarrow th(\beta_C I_A)$ . It gives us a way to transform corresponding equations determining  $T_C$  from Sec. II A to the case of classical Heisenberg spins in frame of the same model approach. Let us also mention that limits  $\lim_{I_A \rightarrow \infty} \mathcal{L}(\beta_C I_A S^2) = 1$  and  $\lim_{I_A \rightarrow \infty} th(\beta_C I_A) = 1$  coincide in magnitude, while for finite values of  $I_A$  values of  $th(\beta_C I_A)$  and  $\mathcal{L}(\beta_C I_A S^2)$  are slightly different (if  $S = 1$  is assumed). In the Table II values of  $T_C$  are listed for the case of  $S = 1$  for different strength of exchange coupling within dimer  $I_A$ . From Fig. 5 one can see that values of  $T_C$  vary not more than 10% from  $T_C(I_A = I_B)$  when  $0 < I_A < \infty$ . Again with increase of number of atoms in elementary magnetic cell  $T_C$  is getting closer and closer to  $T_C(I_A = I_B)$ .

### III. INFLUENCE OF DUMBBELL EXCHANGE STRENGTH ON $T_C$ OF THE RHOMBOHEDRAL PHASE OF $\text{Gd}_2\text{Fe}_{17}$

Here we examine influence of strongly exchange coupled dumbbell Fe atoms on  $T_C$  of the rhombohedral phase of  $\text{Gd}_2\text{Fe}_{17}$ . In this compound there are 17 magnetic

TABLE II: Curie temperature value for different magnetic elementary cells with classical Heisenberg spins with selected strong exchange bond  $I_A$  as a function of ratio  $I_A/I_B$  for  $S = 1$ .  $T_C$  is given in units  $I_B$ .

Magnetic elementary cell	Magnetic atoms number	$I_A=0$	$I_A=I_B$	$I_A=5I_B$	$I_A=10I_B$	$I_A \rightarrow \infty$
$2a \times 2a$	4	1.187	1.333	1.489	1.547	1.609
$3a \times 3a$	9	1.273	1.333	1.389	1.407	1.447
$4a \times 4a$	16	1.302	1.333	1.372	1.386	1.400
$2a \times 2a \times 2a$	8	1.928	2	2.144	2.215	2.299
$3a \times 2a \times 2a$	12	1.954	2	2.101	2.152	2.214
$3a \times 3a \times 2a$	18	1.970	2	2.068	2.104	2.147
$3a \times 3a \times 3a$	27	1.976	2	2.056	2.085	2.121

(Fe) atoms in the elementary magnetic cell, which occupies four types of nonequivalent crystallographic positions. Correspondingly they have different surroundings of neighboring Fe atoms. Namely, for  $Th_2Zn_{17}$ -type rhombohedral structure there are 2 Fe1 atoms in 6c Wychoff positions (dumbbell positions), 3 Fe2 atoms (9d), 6 Fe3 atoms (18f) and 6 Fe4 atoms (18h). Local magnetic moments of different classes of Fe atoms are a bit different from each other:  $\mu_{Fe1} = 2.19\mu_B$ ,  $\mu_{Fe2} = 2.26\mu_B$ ,  $\mu_{Fe3} = 2.17\mu_B$  and  $\mu_{Fe4} = 2.31\mu_B$ .<sup>2</sup> Exchange interaction parameters for the first coordination sphere of different Fe atom classes calculated within the LSDA+U approach<sup>5</sup> in Ref. 2 are presented in Table III.

It was found that largest parameter of exchange interaction  $I_{11}(1) = 287.5$  K couples two Fe1 atoms in the dumbbell position with a distance inbetween  $r_{11}=2.385$  Å. Next largest exchange coupling parameter  $I_{44}(1)=182.2$  K couples magnetic moments of Fe4 atoms with a distance  $r_{44}(1)=2.490$  Å. Having at hand values of exchange interaction constants and local magnetic moments for all Fe magnetic sublattices it is straightforward to estimate  $T_C$  using nearest neighbors mean-field approximation. For this purposes absolute value of spin vector is  $S_i = \mu_{Fe(i)}/2\mu_B$  ( $i=1,2,3,4$ ). To do that first one should define a set of self-consistent equations for average value of  $z$ -component of the spin  $m_i = \langle S_i^z \rangle$ , which are

$$m_i = S_i \mathcal{L}\left(\frac{h_i S_i}{k_B T}\right), \quad (22)$$

where  $h_i$  is Weiss field acting on spin  $S_i$  from nearest neighbors. Next one should linearize right hand sides of Eqs. 22 expanding Langevin function for  $T \rightarrow T_C$

$$m_i = \frac{S_i^2}{3} \frac{h_i}{k_B T}. \quad (23)$$

Taking into account rhombohedral crystal structure of  $Gd_2Fe_{17}$  one can derive set of linear equations for  $m_i$

defining  $T_C$

$$\begin{aligned} m_1 &= \beta_C \frac{S_1^2}{3} [ I_{11}(1)z_{11}(1)m_1 + I_{12}(1)z_{12}(1)m_2 + \\ &\quad + I_{13}(1)z_{13}(1)m_3 + I_{14}(1)z_{14}(1)m_4], \\ m_2 &= \beta_C \frac{S_2^2}{3} [ I_{21}(1)z_{21}(1)m_1 + I_{23}(1)z_{23}(1)m_2 + \\ &\quad + I_{33}(1)z_{33}(1)m_3], \\ m_3 &= \beta_C \frac{S_3^2}{3} [ I_{31}(1)z_{31}(1)m_1 + I_{32}(1)z_{32}(1)m_2 + \\ &\quad + I_{33}(1)z_{33}(1)m_3 + \\ &\quad + (I_{34}(1)z_{34}(1) + I_{34}(2)z_{34}(2))m_4], \\ m_4 &= \beta_C \frac{S_4^2}{3} [ I_{41}(1)z_{41}(1)m_1 + I_{42}(1)z_{42}(1)m_2 + \\ &\quad + (I_{43}(1)z_{43}(1) + I_{43}(1)z_{43}(1))m_3 + \\ &\quad + I_{44}(1)z_{44}(1)m_4], \end{aligned} \quad (24)$$

corresponding exchange integrals  $I_{ij}(1)$  between  $Fe_i$  and  $Fe_j$  crystallographic classes in the first coordination sphere and nearest neighbors number  $z_{ij}(1)$  on a distance  $r_{ij}(1)$  are taken from Table III. At that for cumbersome crystal structures  $z_{ij}(1) \neq z_{ji}(1)$  but  $n_i z_{ij}(1) = n_j z_{ji}(1)$ , where  $n_i$  number of atoms of sort  $Fe_i$  in a elementary magnetic cell.

As was obtained in Ref. 2 Eqs. (24) give  $T_C=429$  K for rhombohedral  $Gd_2Fe_{17}$  which is slightly smaller than experimental one 475 K<sup>1</sup>. A reason of this discrepancy is mainly absence in our model of exchange interactions between Fe and Gd sublattices and oscillating exchange interactions with next coordination spheres. However this result shows that largest part (leading contribution) of  $T_C$  comes from exchange interactions of Fe sublattice between nearest neighbors. Thus it is interesting to explore influence of strongest exchange  $I_{11}(1)$  “in dumbbell” on  $T_C$ .

At the beginning hypothetically one switches off the interaction  $I_{11}(1)$ . Then solution of Eqs. (24) leads to  $T_C(I_{11}(1) = 0)=414$  K which is only 3.5% less than in Ref. 2. Now we consider opposite case  $I_{11}(1) \gg I_{ij}(1)$ , i.e. consider dumbbell pair as dimer. Thus one can apply approach described in Sec. II: Fe1-Fe1 cluster with strong exchange coupling  $I_{11}(1)$  will be treated exactly while

TABLE III: Parameters of exchange in the rhombohedral structure of  $\text{Gd}_2\text{Fe}_{17}$  for the ions of the first coordination sphere.

N	Exchange (K)	Distance (Å)	Number of neighbors	Type
1	$I_{11}(1)=287.5$	$r_{11}(1)=2.385$	$z_{11}(1)=1$	Fe1 (dumbbell) - Fe1 (dumbbell)
2	$I_{44}(1)=182.2$	$r_{44}(1)=2.490$	$z_{44}(1)=2$	Fe4 (corrugated plane) - Fe4 (corrugated plane)
3	$I_{34}(1)=125.9$	$r_{34}(1)=2.551$	$z_{34}(1)=z_{43}(1)=2$	Fe3 - Fe4 (corrugated plane)
4	$I_{24}(1)=121.0$	$r_{24}(1)=2.448$	$z_{24}(1)=4, z_{42}(1)=2$	Fe2 (corrugated layer) - Fe4 (corrugated layer)
5	$I_{34}(2)=105.7$	$r_{34}(2)=2.613$	$z_{34}(2)=z_{43}(2)=2$	Fe3 - Fe4 (corrugated layer)
6	$I_{14}(1)=88.2$	$r_{14}(1)=2.639$	$z_{14}(1)=3, z_{41}(1)=1$	Fe1 (dumbbell) - Fe4 (corrugated layer)
7	$I_{23}(1)=87.1$	$r_{23}(1)=2.423$	$z_{23}(1)=4, z_{32}(1)=2$	Fe3 (dumbbell) - Fe2 (corrugated layer)
8	$I_{12}(1)=83.6$	$r_{12}(1)=2.602$	$z_{12}(1)=3, z_{21}(1)=2$	Fe1 (dumbbell) - Fe2 (corrugated layer)
9	$I_{13}(1)=74.1$	$r_{13}(1)=2.740$	$z_{13}(1)=6, z_{31}(1)=2$	Fe1 (dumbbell) - Fe3
10	$I_{33}(1)=-36.5$	$r_{33}(1)=2.466$	$z_{33}(1)=2$	Fe3 - Fe3

other exchange bonds within mean-field approach. After usual linearization of equations on  $m_1 = \langle S_1^z \rangle$  for small values of  $m_i$  ( $i = 1, 2, 3, 4$ ) close to  $T_C$  one gets equation

$$m_1 = \beta_C \frac{S_1^2}{3} [1 + \mathcal{L}(\beta_C I_{11}(1) S_1^2)] \times \quad (25)$$

$$\times [I_{12}(1) z_{12}(1) m_2 + I_{13}(1) z_{13}(1) m_3 +$$

$$+ I_{14}(1) z_{14}(1) m_4],$$

which should substitute the first equation of Eqs. (24).

For the rhombohedral phase of  $\text{Gd}_2\text{Fe}_{17}$  second largest exchange parameter is  $I_{44}(1)=182.2$  K. Let us take  $I_{11}(1) = 10I_{44}(1)=1822$  K then  $T_C(I_{11}(1) = 10I_{44}(1))=456$  K. And finally for the limit  $I_{11}(1) \rightarrow \infty$ , when  $\mathcal{L}(\beta_C I_{11}(1) S_1^2) \rightarrow 1$ , Curie temperature is  $T_C(I_{11}(1) \rightarrow \infty)=464$  K. Thereby even dumbbell exchange  $I_{11}(1) \rightarrow \infty$  gives rise of  $T_C$  only 10%. Also from Eqs. (26) one follows that for  $I_{11}(1) \rightarrow \infty$  between spins  $S_1$  and  $S_2$  Weiss field acting from other spins of atoms Fe2, Fe3 and Fe4 is doubled.

Another interesting observation one can do for Curie point  $T_C$  calculated from averaged over all types of Fe atoms values of exchange constant  $\bar{I}$ , spin value  $\bar{S}$  and number of nearest neighbors  $\bar{z}$

$$k_B T_C = \frac{1}{3} \bar{I} (\bar{S})^2 \bar{z}, \quad (26)$$

$$\bar{S} = \sum_{i=1}^4 n_i S_i / \sum_{i=1}^4 n_i = 1.12,$$

$$\bar{z} = \sum_{i=1,j}^4 n_i z_{ij} / \sum_{i=1}^4 n_i = 10,$$

$$\bar{I} = 3k_B T_C / (\bar{S})^2 \bar{z} = 102.6 \text{ K},$$

here  $T_C=429$  K. From Table III one can see that most of values of exchange integrals are very close to the value of  $\bar{I}$ . Thus these Fe-Fe exchange bonds are responsible for the Curie point value rather than Fe1-Fe1 exchange bond only, independently how strong it is.

#### IV. CONCLUSION

In this work we proposed description of several model magnetic structures with diluted magnetic dimers which do not form any infinite magnetic cluster. The selected strong exchange bond  $I_A$  is supposed to be much larger than other exchange interactions  $I_B$  in the elementary magnetic cell. Following ideas of Ref. 3 the magnetic dimer is treated exactly while other couplings are treated within mean-field approximation. In contrast to regular mean-field approximation latter approach allows one to obtain finite Curie temperatures  $T_C$  even for  $I_A \rightarrow \infty$  which is physically correct. Also if was demonstrated that for  $0 \leq I_A/I_B < \infty$   $T_C$  deviates just about  $\pm 10\%$  from the value  $T_C(I_A = I_B)$  and is getting closer to that while number of atoms in the elementary magnetic cell grows. For the case  $I_A \rightarrow \infty$  we obtained doubling of spin value of spins forming a dimer, which corresponds to doubling of Weiss field acting on the spin from other spins in the elementary magnetic cell. After all one can conclude that such diluted magnetic dimers do not influence very much on  $T_C$  value of the whole system in the case  $I_A \rightarrow \infty$ , and then even less for finite  $I_A$ .

Based on these results we perform analogous calculations of  $T_C$  for real system – the rhombohedral phase of  $\text{Gd}_2\text{Fe}_{17}$ . There is so called dumbbell Fe1-Fe1 dimer with the largest exchange interaction value  $I_{11}(1)$  in the system. We showed in this investigation that for such complicated crystal structure  $T_C$  is mainly defined by weaker exchange interactions of Fe1 with other Fe atoms in the elementary magnetic cell rather than by the  $I_{11}(1)$  exchange only.

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